

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION
(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents
 United States Patent and Trademark
 Office
 Box PCT
 Washington, D.C.20231
 ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 04 October 2000 (04.10.00)
International application No. PCT/US00/04754
International filing date (day/month/year) 25 February 2000 (25.02.00)
Applicant NAKATA, Kazuyuki

Applicant's or agent's file reference
AD6705PCT

Priority date (day/month/year)
25 February 1999 (25.02.99)

1. The designated Office is hereby notified of its election made:

in the demand filed with the International Preliminary Examining Authority on:

30 August 2000 (30.08.00)

in a notice effecting later election filed with the International Bureau on:

2. The election was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Olivia TEFY Telephone No.: (41-22) 338.83.38
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PATENT COOPERATION TREATY

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From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

JUN 12 2001

To:

Evans, Craig H.
E.I. DU PONT DE NEMOURS AND COMPANY
Legal Patent Records Center
1007 Market Street
Wilmington, Delaware 19898
ETATS-UNIS D'AMERIQUE

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PATENT RECORDS
CENTER

NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL PRELIMINARY
EXAMINATION REPORT
(PCT Rule 71.1)

Date of mailing
(day/month/year) 31.05.2001

Applicant's or agent's file reference

AD6705

IMPORTANT NOTIFICATION

International application No.
PCT/US00/04754

International filing date (day/month/year)
25/02/2000

Priority date (day/month/year)
25/02/1999

Applicant

E.I. DU PONT DE NEMOURS AND COMPANY et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



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REY NOTED



TENT COOPERATION TREATY
PCT
INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Article 36 and Rule 70)

Applicant's or agent's file reference AD6705	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/US00/04754	International filing date (day/month/year) 25/02/2000	Priority date (day/month/year) 25/02/1999
International Patent Classification (IPC) or national classification and IPC C08F8/44		
Applicant E.I. DU PONT DE NEMOURS AND COMPANY et al.		
<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p><input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of 4 sheets.</p> <p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> I <input checked="" type="checkbox"/> Basis of the report II <input type="checkbox"/> Priority III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability IV <input type="checkbox"/> Lack of unity of invention V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement VI <input type="checkbox"/> Certain documents cited VII <input checked="" type="checkbox"/> Certain defects in the international application VIII <input type="checkbox"/> Certain observations on the international application 		
Date of submission of the demand 30/08/2000	Date of completion of this report 31.05.2001	
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Krail, G Telephone No. +49 89 2399 8302 	

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/US00/04754

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*);
Description, pages:

2-8	as originally filed	
1,1a	with telefax of	17/04/2001

Claims, No.:

1-12	with telefax of	17/04/2001
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2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- the language of publication of the international application (under Rule 48.3(b)).
- the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- contained in the international application in written form.
- filed together with the international application in computer readable form.
- furnished subsequently to this Authority in written form.
- furnished subsequently to this Authority in computer readable form.
- The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- the description, pages:
- the claims, Nos.:
- the drawings, sheets:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

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5. This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N) Yes: Claims 1-8,11,12
 No: Claims 9,10

Inventive step (IS) Yes: Claims 1-8,11,12
 No: Claims

Industrial applicability (IA) Yes: Claims 1-12
 No: Claims

**2. Citations and explanations
see separate sheet**

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US00/04754

Re it m V

Reference is made to the following documents:

- D1: GB-A-2 269 822 (OXY-WAX LTD.) 23 February 1994 (1994-02-23)
- D2: GB-A-1 559 048 (ASAHI-DOW LTD.) 16 January 1980 (1980-01-16)
- D3: US-A-3 674 896 (C. L. PURCELL) 4 July 1972 (1972-07-04)
- D4: BE 657 147 A (UCC) 1 April 1965 (1965-04-01)

1.) Citations

D1 GB 2 269 822

Aqueous emulsion of an ethylene / (10 -25 %) methacrylic acid copolymer; completely neutralized with ammonium hydroxide (claims 1 + 3; page 2, para 3-4): for coating substrates (p. 2, para 1,2).

D2 GB 1 559 048

Ethylene/ (1-25 %) methacrylic acid copolymer (claims 1 + 3) " at least partially neutralized" (claim 1) with ammonia (claim 1) excellent film forming properties and suitable for coating various substrates (p.3, l. 31-35, 52 et seq.)

D3 US 3 674 896

During the drying step ammonia is evolved leaving the ethylene acrylic polymer on the substrate (abstract).

D4 BE 657 147

Ethylene /acrylic acid ((2-30%, claim 5) is transferred into a stable emulsion by 30-100, preferably 40-60 mole % of a base, e.g. ammonia (claim 10). For coating substrates (p. 1, 2)

Claim 1 of the application reads "*consisting of*". Thus, the following citations are less relevant.

Derwent Abstract of JP 10 147683

aqueous dispersion of an ethylene/acrylic acid (>10 wt%) copolymer and ammonia in amount to neutralise 70 - 100 % of the carboxyl groups *and a cyclic diamine*.

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US00/04754

CH 476 041

To a ethylene/ methacrylic acid (5-75 %) copolymer is added a base in an amount equivalent to 20 -100 % of the carboxyl groups *in the presence of a solvent* (claim 3): stable aqueous latices without emulsifiers.

Ammonia is less preferred (3/39)

2.) Process claims 11 and 12 and product claims 1 - 8 directed to the aqueous dispersion

It was known in the art to provide stable aqueous dispersions of ethylene / methacrylic acid copolymers by the use of ammonia alone, cf. D1 and D2.

According to D2 the methacrylic acid moieties are partially neutralised, according to D1 they are completely neutralised.

According to the application, ammonia in an amount required for neutralizing 110 - 150 % of the carboxyl groups is present.

Thus, novelty is given with the distinguishing feature being 110 -150 % instead of 100% ammonia.

The problem to use ammonia alone in order to provide stable emulsions and the problem to provide therefrom coating free of alkali was solved by D1 and D2 already.

In support of inventive step the application contains comparative experiments which are intended to show that by the increase of the ammonia ratio to 110-150 % the dispersion remains free of non-dispersed particles for an extended period of time.

Apparently this was beyond expectation. Thus inventive step can be acknowledged.

3.) The coatings obtained from the aqueous dispersions, Claims 9 and 10

The fact that to claim product claim 1 is referred to, does not change the fact that they are independent claims.

In this Office acting as the International Preliminary Examination Authority claims for products defined in terms of a process of manufacture are considered to comply with

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US00/04754

the requirements of Art. 33 (2) and (3) PCT only if the products as such - irrespective of the method or process in which they were produced - fulfil these requirements, i.e. they are new and inventive. A product is deemed not to be rendered novel merely by the fact that it is produced by means of a new process. Rather it has to be shown that distinct differences exist in the properties of the products.

Furthermore in this Office acting as the International Preliminary Examination Authority another degree of impurities, here ammonia, normally is not considered as to impart novelty to a known chemical product.

During the drying step of the coated substrates ammonia is evolved leaving the ethylene acrylic polymer on the substrate (cf. D3, abstract).

Thus, on the face of it, the products obtained D1, D2 and application are identical. If the applicant nevertheless argues against plausibility that his product is novel, the burden of proof is with him.

Re item VII

Claim 1 of D2 reads "at least partially neutralized with . . . metallic and / or ammonium ions". Apparently the acknowledgement of D1 (page 1 of the application documents) is not conform to this (Rule 5.1(a)(ii) PCT).

TITLE**AQUEOUS DISPERSION COMPOSITION AND MANUFACTURING
METHOD FOR THE COMPOSITION****FIELD OF THE INVENTION**

The present invention relates to an aqueous dispersion composition having good stability and dispersion properties and a process for making it by dispersing an ethylene-methacrylic acid copolymer in water using an amount of ammonia in excess of amount that would be needed to neutralize methacrylic acid. It also relates to the application of this aqueous dispersion in making coated substrates.

BACKGROUND OF THE INVENTION

Aqueous dispersion compositions of ethylene α,β -ethylenically unsaturated carboxylic acid copolymers such as ethylene-acrylic acid copolymer or ethylene-methacrylic acid copolymer are known and sold commercially. They can be easily made by dispersing the readily available ethylene acid copolymers in water using an alkali metal compound and are useful in various applications such as coating film. However, because the film coated with such dispersion compositions has a poor waterproofness, it cannot be used for applications requiring a film that is waterproof.

It is known that an ethylene-acrylic acid copolymer can be used as an excellent raw material for an aqueous dispersion composition obtained using ammonia as dispersion aide. Although it has been difficult to obtain an aqueous dispersion composition using ammonia alone, a film coated with such aqueous dispersion should have a good waterproofness since there is no alkali ion to absorb water.

Uniformly dispersing ethylene-methacrylic acid copolymer in water using a mixture of ammonia and an alkali metal has been known to be difficult. Such aqueous dispersions of ethylene-methacrylic acid copolymer could be obtained by adding small amounts of a surfactant as a supplemental disperser. However, because the coated film using such dispersions have both poor waterproofness and bleed-out, causing contamination, it was not preferred.

An object of this invention is to find a way to use ammonia alone as a dispersion aide to make a uniform aqueous dispersion composition of an ethylene-methacrylic acid copolymer that is stable for a long time, that is has a good shelf-life of say a year or more. Another object was to find a way to make a film (or other substrate) that when coated with such an aqueous dispersion would have good waterproofness.

5 **WHAT IS CLAIMED IS:**

1. A stable, uniform aqueous dispersion consisting essentially of a dispersion in water of component (A), an ethylene-methacrylic acid copolymer containing 15-35 wt% methacrylic acid, and component (B), ammonia in an amount required for neutralizing 110-150% of the carboxyl groups of component (A).
10
2. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid contains 18-30 wt% methacrylic acid.
15
3. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid contains 15-25 wt% methacrylic acid.
15
4. The aqueous dispersion of claims 1, 2, or 3 wherein the ammonia is present in an amount sufficient to neutralize 120-140% of the carboxyl groups.
20
5. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid copolymer comprises 5-50 wt% of the dispersion and preferably has a melt flow rate of 50-2000 grams/10 minutes at 190°C/2160 gram load.
25
6. The aqueous dispersion of claim 3 wherein the ethylene-methacrylic acid copolymer comprises 5-50 wt% of the dispersion and preferably has a melt flow rate of 50-2000 grams/10 minutes at 190°C/2160 gram load.
30
7. The aqueous dispersion of claim 4 wherein the ethylene-methacrylic acid copolymer has a melt flow rate of 60-1500 grams/10 minutes at 190°C/2160 gram load.
35
8. The aqueous dispersion of claim 5 wherein the ethylene-methacrylic acid copolymer has a melt flow rate of 60-1500 grams/10 minutes at 190°C/2160 gram load.
35
9. A coated substrate obtained by applying the aqueous dispersion of claim 1, 2 or 3 to the substrate for coating, then drying to form a coated substrate.
40
10. The coated substrate wherein the substrate is a film.

- 5 11. A process for making a stable, uniform aqueous dispersion of ethylene-methacrylic acid consisting essentially of mixing an ethylene-methacrylic acid copolymer containing 15-35 wt% methacrylic acid in water in the presence of sufficient ammonia to neutralize 110 to 150% of the carboxylic acid groups in the ethylene-methacrylic acid copolymer for a sufficient time to uniformly disperse
10 the ethylene-methacrylic acid copolymer in the water.
12. The process of claim 11 wherein the mixing is carried out at a temperature of about 90 to about 150°C for about 10 minutes to about 2 hours.